Pd Nanocrystals Encapsulated in Ni-Metal-organic Framework-derived Hollow Nanostructure Carbon for Efficient Thermal CO Oxidation: Unveiling the Effect of Porosity

Adewale K. Ipadeola[a,b,ǂ], Ahmed Gamal[a,b,c,ǂ], Aboubakr M. Abdullah[a,*], Kenneth I. Ozoemena[c] and Kamel Eid[b,*]

[a.]Center for Advanced Materials, Qatar University, Doha 2713, Qatar.

[b.]Gas Processing Center(GPC), College of Engineering, Qatar University, Doha 2713, Qatar.

[c.]Molecular Science Institute, School of Chemistry, University of the Witwatersrand, Private Bag 3, PO Wits, Johannesburg, South Africa

ǂ Contributed equally to this work

*Corresponding authors: bakr@qu.edu.qa; kenneth.ozoemena@wits.ac.za; kamel.eid@qu.edu.qa

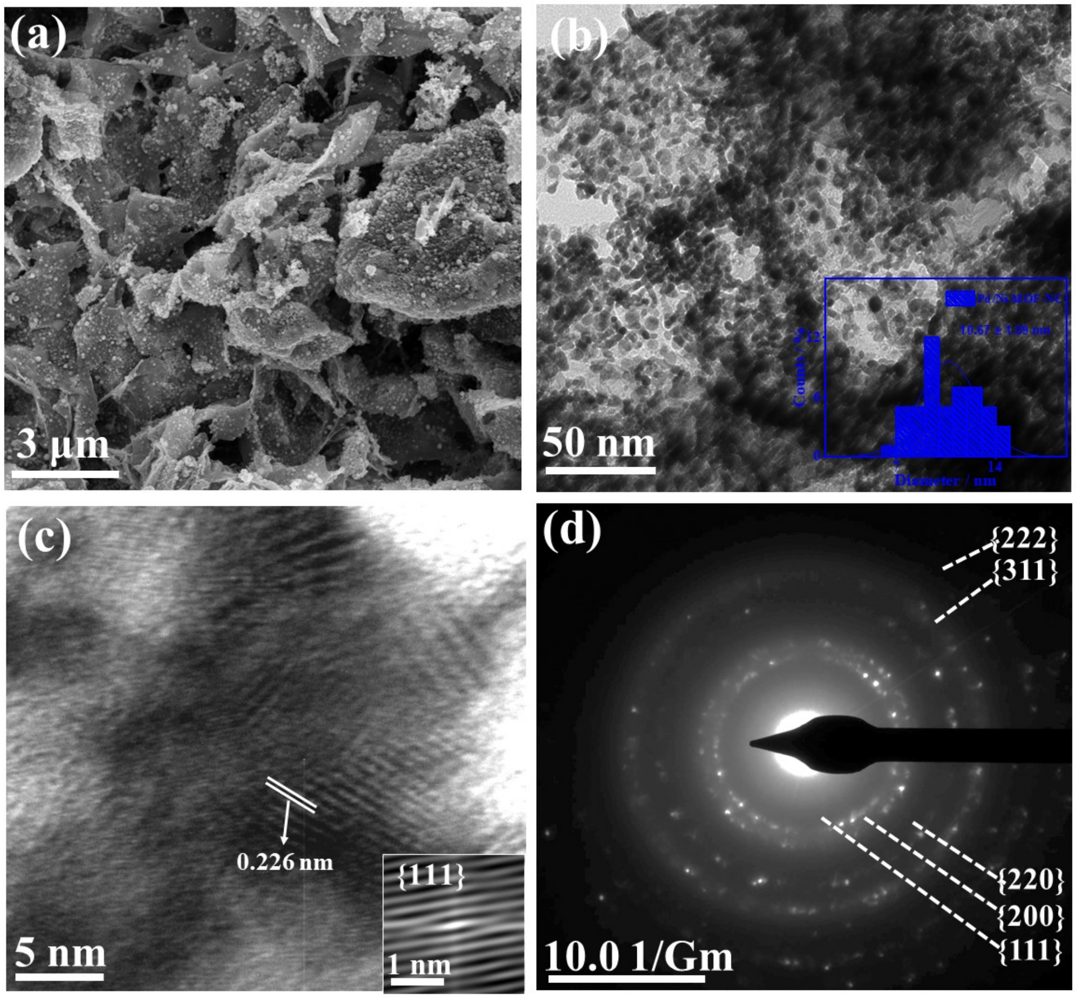

Fig. S1. (a) SEM, (b) TEM with nanoparticle size distribution, (c) HRTEM, and (d) SAED of Pd/Ni-MOF-NC.

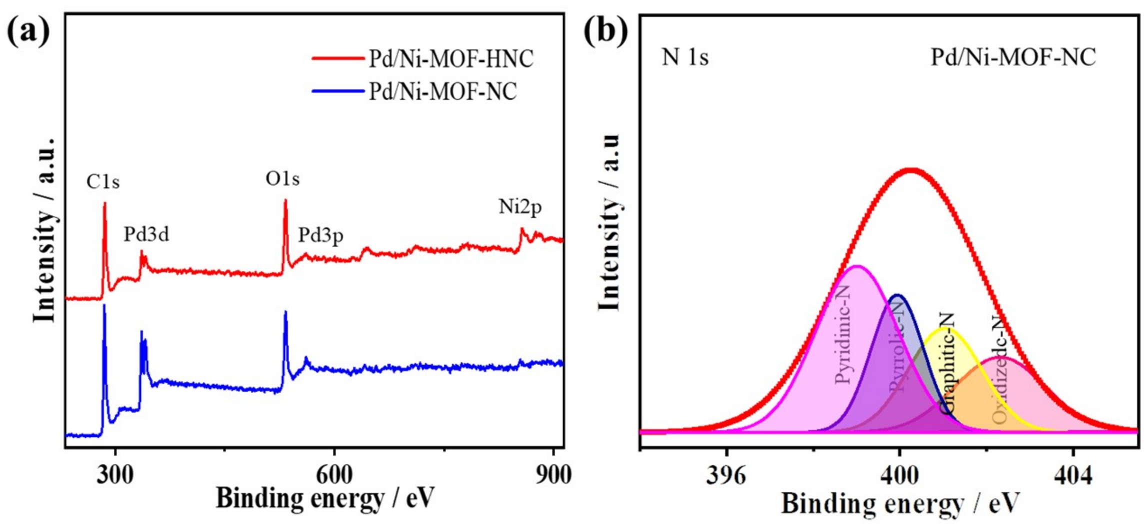

Fig. S2. (a) XPS full-scan of Pd/Ni-MOF-HNC and Pd/Ni-MOF-NC, and (b) high-resolution N 1s of Pd/Ni-MOF-NC.

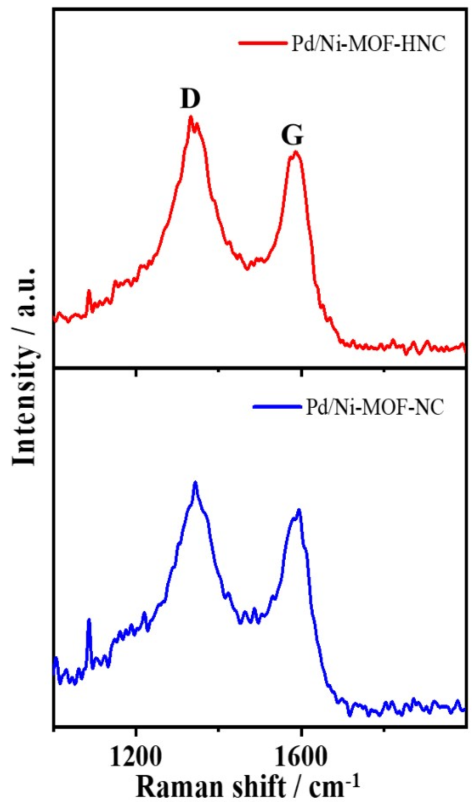

Fig. 3. Raman of Pd/Ni-MOF-HNC and Pd/Ni-MOF-NC.

Table S1. The comparison of low-temperature CO oxidation activity of the as-synthesized catalysts with Pd-based catalysts in the literature.

Catalysts	Preparation methods	Morphology	T_{100} (^{o}C)	Refs
Pd/Ni-MOF-HNC	Microwave-irradiation	Porous nanosheets	114.4	This work
Pd/Ni-MOF-NC	Microwave-irradiation	Nanosheets	153.8	This work
Pd/CeSn75-800	Counter precipitation/ calcination	Core-shell	~ 100	[1]
Pd@SiO_2/TiO_2-500	Precipitation/ calcination	Core-shell	~ 400	[2]
CeO_2-Pd/S-800-5h	Impregnation/ calcination	2d hexagonal mesopores	~ 75	[3]

Pd_1@HEFO Pd@CeO_2	Ball milling/ annealing/etching	Cubic	170 253	[4]
Pd/MgO(5)-h-BN	Impregnation/ calcination	Nanosheets	140	[5]
$Ce_{1-x}Pd_xO_{2-\delta}$ (PC3)	hydrothermal/reduction/calcination	Nanocrystals	~ 95	[6]
Pd/MgAl-HT	Deposition-precipitation	Nanocrystals	~ 90	[7]
Pd-1%P	Wet- impregnation/ calcination	Fiber-like lamellar	~ 270	[8]
LaAlPd(0.8)O_3-600	Impregnation/ Calcination	Perovskite	~ 325	[9]
Pd@SiO_2-673	Polymerization/calcination	Core-shell	~ 130	[10]
$Pd_{0.83}Co_{0.17}$/C	Wet impregnation	Nanocrystals	150	[11]
$Pd_{0.5}$/CeHfZrSnErO_x $Pd_{1.0}$/CeHfZrSnErO_x	Ultrasound-mediated co-precipitation	Cubic	140 150	[12]
Pd-SSZ-13	Ion exchange/ calcination	Cubic particles	~ 175	[13]
PdO_x/CeO_2	Radio frequency sputtering	Dendrite-like	250	[14]
4%Pd/R-CeO_2	Impregnation/ annealing	Rod, cubic and octahedral	50	[15]
Pd-Cu/gC_3N_4NWs	Protonation/annealing	Nanowires	149	[16]
Pd/Cu/gC_3N_4NTs	Protonation/annealing	Nanotubes	154	[17]
Au/Pd/gC_3N_4NFs	Protonation/annealing	Nanofibers	144	[18]
Pd- impeded 3D porous graphene (3D Pd-E-PG)	Low-power microwave radiation	3D porous nanosheets	190	[19]

$AuPd/TiO_2$	Incipient wetness method	Nanospheres	190	[20]

MOF-derived hollow N-doped carbon (MOF-HNC) nanostructures, MOF-derived N-doped carbon (MOF-NC) nanostructure, high-entropy fluorite oxide (HEFO), hexagonal boron nitride (h-BN), calcination at 500 ^{o}C for 18 h (PC3), hydrotalcite-like (HT), SSZ-13 zeolites (SSZ-13), rod-like (R)

References

1. O. A. Stonkus, A. V. Zadesenets, E. M. Slavinskaya, A. I. Stadnichenko, V. A. Svetlichnyi, Y. V. Shubin, S. V. Korenev and A. I. Boronin, *Catal. Commun.*, 2022, **172**, 106554.
2. A. Takabayashi, F. Kishimoto, H. Tsuchiya, H. Mikami and K. Takanabe, *Nanoscale Adv.*, 2023, **5**, 1124-1132.
3. P. R. Murthy, S. Munsif, J.-C. Zhang and W.-Z. Li, *Industrial & Engineering Chemistry Research*, 2021, **60**, 14424-14433.
4. H. Xu, Z. Zhang, J. Liu, C.-L. Do-Thanh, H. Chen, S. Xu, Q. Lin, Y. Jiao, J. Wang and Y. Wang, *Nat. Commun.*, 2020, **11**, 3908.
5. L. Li, X. Liu, H. He, N. Zhang, Z. Liu and G. Zhang, *Catal. Today*, 2019, **332**, 214-221.
6. J. Ye, Y. Xia, D.-g. Cheng, F. Chen and X. Zhan, *Int. J. Hydrog. Energy*, 2019, **44**, 17985-17994.
7. X. Lin, J. Zhou, Y. Fan, Y. Zhan, C. Chen, D. Li and L. Jiang, *Dalton Trans.*, 2018, **47**, 14938-14944.
8. J. Dong, J. Wang, J. Wang, M. Yang, W. Li and M. Shen, *Catal. Sci. Technol.*, 2017, **7**, 5038-5048.
9. Y. J. Kim, J. H. Lim, B. K. Cho, S. B. Hong, I.-S. Nam and J. W. Choung, *J. Catal.*, 2015, **330**, 71-83.
10. Y. Xu, J. Ma, Y. Xu, L. Xu, L. Xu, H. Li and H. Li, *RSC Adv.*, 2013, **3**, 851-858.
11. I. B. Aragão, F. R. Estrada, D. H. Barrett and C. B. Rodella, *Mol. Catal.*, 2022, **526**, 112377.
12. F. Okejiri, J. Fan, Z. Huang, K. M. Siniard, M. Chi, F. Polo-Garzon, Z. Yang and S. Dai, *iscience*, 2022, **25**, 104214.
13. D. Chen, H. Lei, W. Xiong, Y. Li, X. Ji, J.-Y. Yang, B. Peng, M. Fu, P. Chen and D. Ye, *ACS Catal.*, 2021, **11**, 13891-13901.
14. L. S. Kibis, A. A. Simanenko, A. I. Stadnichenko, V. I. Zaikovskii and A. I. Boronin, *J. Phys. Chem. C*, 2021, **125**, 20845-20854.
15. X. Zhang, W. Li, Z. Zhou, K. Chen, M. Wu and L. Yuan, *Mol. Catal.*, 2021, **508**, 111580.
16. K. Eid, Y. H. Ahmad, A. T. Mohamed, A. G. Elsafy and S. Y. Al-Qaradawi, *Catalysts*, 2018, **8**, 411.
17. K. Eid, M. H. Sliem, K. Jlassi, A. S. Eldesoky, G. G. Abdo, S. Y. Al-Qaradawi, M. A. Sharaf, A. M. Abdullah and A. A. Elzatahry, *Inorg. Chem. Commun*, 2019, **107**, 107460.
18. K. Eid, M. H. Sliem, A. S. Eldesoky, H. Al-Kandari and A. M. Abdullah, *Int. J. Hydrog. Energy*, 2019, **44**, 17943-17953.
19. R. Kumar, J.-H. Oh, H.-J. Kim, J.-H. Jung, C.-H. Jung, W. G. Hong, H.-J. Kim, J.-Y. Park and I.-K. Oh, *ACS nano*, 2015, **9**, 7343-7351.
20. A. Teixeira-Neto, R. Gonçalves, C. Rodella, L. Rossi and E. Teixeira-Neto, *Catalysis Science & Technology*, 2017, **7**, 1679-1689.